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## PATENT ABSTRACTS OF JAPAN

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(54) POSITIVE-ELECTRODE ACTIVE MATERIAL FOR LITHIUM SECONDARY BATTERY  
AND ITS MANUFACTURE

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a positive-electrode active material for a lithium secondary battery superior in electrochemical performance, and to provide a manufacturing method of the positive-electrode active material for a lithium secondary battery capable of suppressing generation of an undesired impurity phase (minor phase).

SOLUTION: A positive-electrode active material for a lithium secondary battery is constituted of a substance of the formula  $\text{Li}_x\text{Ni}_{1-y}\text{Co}_y\text{O}_2$ , including giant particles having a size of 1 to 25  $\mu\text{m}$  formed of a multiplicity of fine particles having a size of 0.4 to 0.7  $\mu\text{m}$ . A lithium salt, nickel salt and cobalt salt are dissolved in a solvent at a molar ratio of (0.95-1.06):(0.5-1):(0-0.5), then a chelator is added thereto, a gel is manufactured by heating this mixture, an organic/inorganic

precursor is formed by heat-decomposing the gel, and the positive electrode active material of the formula for a lithium secondary battery is manufactured by heat-treating the precursor. In the formula, (x) is 0.95 to 1.06, more preferably, 1.01 to 1.05, and (y) is 0 to 0.5.

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#### DETAILED DESCRIPTION

[Detailed Description of the Invention]  
[0001]

[The technical field to which invention belongs] this invention relates to the manufacture method of the positive active material for lithium secondary batteries which can start the positive active material for lithium secondary batteries, and its manufacture method, can adjust the capacity of the positive active material for lithium secondary batteries

whose electrochemical property improved in more detail, and a positive active material, and can be detailed and can manufacture an active material particle uniformly.

[0002]

[Description of the Prior Art] Recently, a miniaturization and lightweight-izing of electronic equipment are realized, and research on the lithium secondary battery which has an energy density high as a power supply of portable electronic equipment is briskly advanced in connection with use of portable electronic equipment becoming common.

[0003] A lithium secondary battery uses the matter in which the intercalation (intercalation) and day intercalation (deintercalation) of a lithium ion are possible for a negative electrode and a positive electrode, is filled up with the movable organic electrolytic solution or the polymer electrolytic solution of a lithium ion between the aforementioned positive electrode and a negative electrode, and is manufactured. The aforementioned lithium secondary battery generates electric energy by oxidization in case the intercalation / day intercalation of the lithium ion are carried out by the aforementioned positive electrode and the negative electrode, and the reduction reaction.

[0004] Although the lithium metal might be used as a negative-electrode (anode) active material of such a lithium secondary battery, since a dendrite (dendrite) is formed on the front face of a lithium metal into the charge-and-discharge process of a cell when using a lithium metal, there is danger of a cell short circuit and cell explosion. A lithium ion can be accepted in reversible, or it can supply, maintaining structure and an electrical property, in order to solve such a trouble, and the carbon system matter with which half cell potential (half cell potential) was similar with the lithium metal is widely used as a negative-electrode active material at the time of insertion of a lithium ion, and desorption.

[0005] generally the chalcogen ghost (chalcogenide) of the metal in which insertion and desorption of a lithium ion are possible as a positive-electrode (cathode) active material of a lithium secondary battery uses it -- having -- as a typical thing --  $\text{LiCoO}_2$  and  $\text{LiMn}_2\text{O}_4$ ,  $\text{LiNiO}_2$ , and  $\text{LiNi}_{1-x}\text{Co}_x\text{O}_2$  (compound metallic oxides, such as  $0 < x < 1$  and  $\text{LiMnO}_2$ , are put in practical use.) Among the aforementioned positive active materials, although it is widely used since it has the demerit in which capacity is small and  $\text{LiCoO}_2$  has the electrical conductivity of about  $10^{-2} \sim 1 \text{ S/cm}$ , a high cell voltage, and the outstanding electrode characteristic at a room temperature, although it has the advantage in which there is also little environmental pollution while Mn system active materials, such as  $\text{LiMn}_2\text{O}_4$  and  $\text{LiMnO}_2$ , are easy to compound and a price is comparatively cheap, the stability at the time of high charge and discharge is low, and it has the trouble that a Moreover, although  $\text{LiNiO}_2$  has the cheapest price among the aforementioned positive active materials

and service capacity and charge capacity are large, it has the demerit in which composition is difficult.

[0006] Generally, such a compound metallic oxide is manufactured by the solid-phase-reaction method for mixing the raw material powder of a solid state and calcinating this. For example, after mixing and heat-treating the hydroxide containing nickel (OH)<sub>2</sub>, Co (OH)<sub>2</sub>, or nickel and Co, the method of manufacturing LiNi<sub>1-x</sub>Co<sub>x</sub>O<sub>2</sub> (0 < x < 1) through process, such as trituration and grain-size judgment, is indicated by JP, 8-153513, A (SONY). After making LiOH, nickel oxide, and Co oxide react, sintering this primarily at 400-580 degrees C as other methods and forming an initial oxide, there is a method of sintering secondarily at 600-780 degrees C, and manufacturing a perfect crystalline active material.

[0007]

[Problem(s) to be Solved by the Invention] However, the method of manufacturing the aforementioned conventional compound metallic oxide has synthetic, comparatively high temperature, and it is difficult for it to adjust physical properties generated while the size of the particle of a reactant is comparatively large, such as a particle shape (Morphology) of an active material and a surface characteristic (a surface area, size of pore). Since the physical property of such an active material is an important factor which does influential influence <sup>size</sup> size, in order to maximum-ize the property of a cell in the electrochemical property of a cell, the casting plan which can adjust arbitrarily the physical property which these electrode material has is required.

[0008] this invention is for solving the aforementioned trouble, and the purpose of this invention is by changing the lithium equivalent in a compound metallic oxide, and adjusting physical characteristics, such as a size of a particle, and a surface characteristic, to offer the positive active material for lithium secondary batteries which was excellent in the electrochemical performance. Other purposes of this invention shorten the heat-treatment time which composition takes while making synthetic temperature low, without performing trituration of a particle, and judgment process in a synthetic process of an active material, and it is in offering the manufacture method of the positive active material for lithium secondary batteries which can suppress generation of the impurity phase (minor phase) for which it does not ask.

[0009]

[Means for Solving the Problem] In order to attain the aforementioned purpose, this invention offers the positive active material for lithium secondary batteries of the following chemical formula 1 containing the giant particle which has the size of 1-25 micrometers formed from the very fine particle of a large number which have the size of 0.4-0.7 micrometers. Moreover, this invention offers the manufacture method of the positive active material for lithium secondary batteries of the following chemical

formula 1 including the process which adds a chelating agent, heats the; aforementioned mixture, manufactures gel, pyrolyzes the; aforementioned gel, forms organic - inorganic precursor, and heat-treats the; aforementioned precursor, after dissolving lithium salt, nickel salt, and cobalt salt in a solvent so that a mole ratio may be set to 0.95-1.06:0.5-1:0-0.5.

[Chemical formula 1]

LixNil-yCoyO<sub>2</sub> (in the above-mentioned formula, x is 0.95-1.06 and 1.01-1.05y of x are 0-0.5 more preferably)

[0010] Hereafter, this invention is explained in more detail. The positive active material for lithium secondary batteries of the chemical formula 1 containing the giant particle which has the size of 1-25 micrometers formed from the very fine particle which has many sizes of 0.4-0.7 micrometers depended on this invention is manufactured by the following methods. Lithium salt, nickel salt, and cobalt salt are dissolved in a solvent so that a mole ratio may be set to 0.95-1.06:0.5-1:0-0.5, and metal salting-in liquid is manufactured. Although anything that can be used for manufacturing the positive active material for lithium secondary batteries as the aforementioned lithium salt, nickel salt, and cobalt salt can be used A lithium nitrate (lithium nitrate), lithium acetate (lithium acetate), The lithium salt chosen from the groups which consist of lithium carbonate (lithium carbonate) and lithium hydroxide (lithium hydroxide), The nickel salt and the cobalt nitrate (cobalt nitrate) which are chosen from the groups which consist of a nickel nitrate (nickel nitrate) and nickel acetate (nickel acetate), It is desirable to use the cobalt salt chosen from the groups which consist of cobalt hydroxide (cobalt hydroxide), cobalt carbonate (cobalt carbonate), and cobalt acetate (cobalt acetate). What is chosen as the aforementioned solvent from the groups which consist of distilled water, ethanol, and a methanol can be used. Moreover, lithium salt, nickel salt, and the metal salt chosen from the groups which become not only cobalt salt but the aforementioned metal salting-in liquid from Mg, aluminum, and Mn can also be added further.

[0011] As the aforementioned chelating agent, the organic polymeric material which has a hydrophilic side chain is used. It surveys so that the polymeric material preferably chosen from the groups which consist of polyvinyl alcohol (polyvinyl alcohol), a polyethylene glycol (polyethyleneglycol), a polyacrylic acid (polyacrylic acid), and a polyvinyl butyral (polyvinylbutyral) may be preferably increased 0.25 to 6 times 0.25 to 10 times to the number of mols of the total metal ion of the aforementioned metal salting-in liquid, and it dissolves in distilled water, and a chelation solution is manufactured. The amount of the aforementioned polymeric material calculates the unit molecular weight of a polymeric material as one mol, and it uses it, calculating so that it may increase 0.25 to 10 times to the number of mols of the used metal

ion. Since it is difficult for viscosity to become very large and to compound gel when the phase (phase) of the matter for which it will ask if the aforementioned polymeric material becomes 0.25 or less times to the number of mols of the total metal ion is not formed and it becomes 10 or more times, it is not desirable.

[0012] If the aforementioned metal salting-in liquid and a chelation solution are mixed, the chelate of the aforementioned macromolecule will be carried out to the aforementioned metal ion, and gel will be formed, when the aforementioned metal ion and a macromolecule come to be uniformly distributed in a solution, heat this mixed solution at 100-120 degrees C and evaporate water.

[0013] Subsequently, organic - inorganic-polymer precursor which pyrolyzed the aforementioned gel at 300-400 degrees C for 1 to 5 hours, and the metal ion and the polymeric material have combined is manufactured. this time -- a programming rate -- the maximum -- it can be made late, for example, a temperature up can be carried out the speed for 1-degree-C/ Macromolecule disassembly of a chelation solution may not be carried out for the aforementioned pyrolysis temperature to a low case good from 300 degrees C, and the precursor of uniform composition may not be formed. Moreover, when pyrolysis temperature is higher than 400 degrees C, the target precursor may not be formed but the crystalline substance for which it does not ask may be generated. The aforementioned precursor has the size of the particle below the micrometer (sub-micron) containing carbon.

[0014] The primary aforementioned precursor is heat-treated at the temperature of 700-900 degrees C under air or oxygen atmosphere for 5 to 20 hours, and the positive active material of LixNil-yCoyO2 is manufactured. If a precursor is heat-treated as mentioned above, the active material of a single phase can be manufactured without forming an impurity (minor phase).

[0015] Thus, if the 1st order is heat-treated and an active material is manufactured after mixing lithium salt, cobalt salt, and nickel salt, the giant particle which the very fine particle of a large number which have the size of 0.4-0.7 micrometers gathers, and has the size of 1-25 micrometers will be formed.

[0016] It is more desirable to heat-treat the secondary active material heat-treated [ aforementioned ] the 1st order at the temperature of 400-600 degrees C under air or oxygen atmosphere for 8 to 10 hours, and to manufacture a positive active material. If the secondary compound heat-treated [ aforementioned ] the 1st order is heat-treated, a crystal will come to be stable and the electrochemical property of the cell manufactured by this using this active material will improve. Moreover, if secondary heat treatment is carried out, the size of the particle of the active material manufactured will become smaller.

[0017] Moreover, since the homogeneity of a medium is secured and the

lithium equivalent is changed by using an organic macromolecule as a chelating agent as mentioned above, the size of the particle of an active material, the configuration of a particle, a surface characteristic, etc. can be adjusted arbitrarily.

[0018]

[Embodiments of the Invention] Hereafter, the desirable example and the example of comparison of this invention are indicated. However, it does not pass over the following example in the desirable example of this invention, and this invention is not limited to the following example. (Example 1)  $\text{LiNO}_3$  was dissolved in distilled water, after surveying correctly 0.8 mols, and  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  for one mol and nickel  $(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  to 0.2 mols, and metal salting-in liquid was manufactured. If a metal salt dissolves in distilled water completely, it will become a transparent, a little black solution. After surveying the polyvinyl alcohol which is a polymeric material as a chelating agent so that it may increase 0.5 times to the number of mols of the total metal ion, it dissolved in distilled water and the chelation solution was manufactured. The aforementioned metal salt and the chelation solution were mixed, it heated at the temperature of about 110 degrees C, water was evaporated, and gel was manufactured. After putting this gel into an alumina container, organic-inorganic precursor which heat-treated at about 300 degrees C for 3 hours, and the metal ion and the polymeric material have combined was manufactured. The primary aforementioned precursor was heat-treated at 750 degrees C by dry air atmosphere for 12 hours, and the crystalline substance of  $\text{Li}1.00\text{nickel}0.8\text{Co}0.202$  was manufactured. Carrying out blowing of the dry air for this crystalline substance, the 2nd order was heat-treated at the temperature of 500 degrees C for 10 hours, and the positive active material for lithium secondary batteries was manufactured. Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0019] (Example 2) The positive active material for lithium secondary batteries of  $\text{Li}1.02\text{nickel}0.8\text{Co}0.202$  was manufactured by the same method as the aforementioned example 1 except having used 1.02 mol of  $\text{LiNO}_3$ . Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0020] (Example 3) 1.04 mol of  $\text{LiNO}_3$  were used and the positive-electrode matter for lithium secondary batteries of  $\text{Li}1.04\text{nickel}0.8\text{Co}0.202$  was manufactured by the same method as the aforementioned example 1 except having carried out secondary heat treatment for 8 hours. Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0021] (Example 4) The positive active material for lithium secondary batteries of  $\text{Li}1.06\text{nickel}0.8\text{Co}0.202$  was manufactured by the same method



as the aforementioned example 1 except having used 1.06 mol of  $\text{LiNO}(\text{s})_3$ . Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0022] (Example 5) The positive active material for lithium secondary batteries of  $\text{Li}_{0.95}\text{nickel}_{10.7}\text{Co}_{0.302}$  was manufactured by the same method as the aforementioned example 1 except having used  $[\text{LiNO}_3]$  0.7 mols, and 0.3 mols  $\text{Co}(\text{NO}_3)$  and  $6\text{H}_2\text{O}$  for 0.95 mols and  $\text{nickel}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ . Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0023] (Example 6) The positive active material for lithium secondary batteries of  $\text{Li}_{1.00}\text{nickel}_{10.7}\text{Co}_{0.302}$  was manufactured by the same method as the aforementioned example 5 except having used one mol of  $\text{LiNO}(\text{s})_3$ . Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0024] (Example 7) The positive active material for lithium secondary batteries of  $\text{Li}_{1.04}\text{nickel}_{10.7}\text{Co}_{0.302}$  was manufactured by the same method as the aforementioned example 5 except having used 1.04 mol of  $\text{LiNO}(\text{s})_3$ . Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0025] (Example 8) The positive active material for lithium secondary batteries of  $\text{Li}_{1.06}\text{nickel}_{10.7}\text{Co}_{0.302}$  was manufactured by the same method as the aforementioned example 5 except having used 1.06 mol of  $\text{LiNO}(\text{s})_3$ . Thus, the coin form cell was manufactured to the counter electrode with the manufactured positive active material using the lithium metal.

[0026] Drawing 1 a shows the XRD pattern of  $0.20\text{Li}_x\text{Ni}_{0.8}\text{Co}_2$  crystalline substance which heat-treated the 1st order and was manufactured, changing the amount of a lithium according to an example 1 or an example 4. Moreover, drawing 1 b shows the XRD pattern of  $0.30\text{Li}_x\text{Ni}_{0.7}\text{Co}_2$  crystalline substance which heat-treated the 1st order and was manufactured according to the example 5 or the example 8. All the peaks of the product which the amount of the lithium added was changed to 0.95 mols or 1.06 mols, and was manufactured were the same as shown in drawing 1 a and 1b. Therefore, even if it changes the amount of a lithium, it turns out that the structure of a product where 1.00 mols added and the lithium was manufactured is maintained as it is. \* display in drawing 1 a shows Si base peak.

[0027] Drawing 2 a or 2c is the SEM photograph which expanded the active material precursor manufactured according to examples 1, 2, and 4 by 20000 times, 30000 times, and 30000 times, respectively. As drawing 2 a or 2c shows, it turns out that the active material precursor manufactured according to the example of this invention is formed from the particle below the micrometer.

[0028] Moreover, drawing 3 a or 3c is the SEM photograph which expanded the crystalline substance which heat-treated the 1st order and was manufactured at 750 degrees C according to examples 1, 2, and 4 by 20000

times, 30000 times, and 30000 times, respectively. The product which changed and manufactured the amount of a lithium to 1.00 mols, 1.02 mols, and 1.06 mols according to the examples 1, 2, and 4 of this invention is formed from the giant particle in which many very fine particles gathered for and were formed so that drawing 3 a or 3c may show. As for the giant particle in which the size was uniformly formed in 0.4-0.7 micrometers, a majority of such very fine particles gathered for, and the aforementioned very fine particle was formed, the size is formed in 1-25 micrometers. That is, the active material manufactured by this invention is formed from the giant particle which has the size of 1-25 micrometers in which the very fine particle of a large number which have the size of 0.4-0.7 micrometers gathered for, and was formed.

[0029] Drawing 4 a is the graph which showed the charge-and-discharge property of each coin form cell manufactured as an active material using the crystalline substance which heat-treated the 1st order and was manufactured according to the example 1 (a of drawing 4 a), the example 3 (b of drawing 4 a), and example 4 (c of drawing 4 a) of this invention. Drawing 5 a is the graph which showed the charge-and-discharge property of each coin form cell manufactured using the primary active material which heat-treated the 2nd order and was stabilized according to the example 1 (a of drawing 5 a), the example 3 (b of drawing 5 a), and example 4 (c of drawing 5 a) of this invention. Drawing 4 a and drawing 5 a measure the capacity of a cell, and the potential difference over Li/Li+, carrying out charge and discharge at the rate of 0.1C between 4.3-2.8V. The capacity of the cell manufactured as an active material using the crystalline substance which heat-treated the 1st order and was manufactured so that drawing 4 a and drawing 5 a may show About 184 mAh/g (example 1), about 188 mAh/g (example 3), It was about 147 mAh/g (example 4), and was lower than the capacity (respectively about 187 mAh/g, about 196 mAh/g, about 168 mAh/g) of the cell manufactured using the active material which heat-treated the 2nd order and was manufactured.

[0030] Drawing 4 b Moreover, the example 5 (a of drawing 4 b) of this invention, an example 6 (b of drawing 4 b), It is the graph which showed the charge-and-discharge property of each coin form cell manufactured as an active material using the crystalline substance which heat-treated the 1st order and was manufactured according to the example 7 (c of drawing 4 b), and the example 8 (d of drawing 4 b). Drawing 5 b is the graph which showed the charge-and-discharge property of each coin form cell manufactured using the primary active material which heat-treated the 2nd order and was stabilized according to the example 5 (a of drawing 5 b), the example 6 (b of drawing 5 b), the example 7 (c of drawing 5 b), and example 8 (d of drawing 5 b) of this invention. Drawing 4 b and drawing 5 b also measure the capacity of a cell, and the potential difference over Li/Li+, carrying out charge and discharge at the rate of 0.1C between

4.3-2.8V. Like what was shown in the result shown in drawing 4 b and drawing 5 b at drawing 4 a and drawing 5 a The capacity of the cell manufactured as an active material using the crystalline substance which heat-treated the 1st order and was manufactured About 155 mAh/g (example 5), About 162 mAh/g (example 6), about 166 mAh/g (example 7), It was about 163 mAh/g (example 8), and was lower than the capacity (respectively about 175 mAh/g, about 182 mAh/g, about 185 mAh/g, about 183 mAh/g) of the cell manufactured using the active material which heat-treated the 2nd order and was manufactured.

[0031] Therefore, the direction which heat-treats the 2nd order and manufactures an active material can heat-treat the 1st order, and capacity can be made to increase rather than it manufactures an active material so that drawing 4 a and 4b, drawing 5 a, and 5b may show. Moreover, even if it compounds a compound metallic oxide so that a lithium may become 0.95-1.06 mols, it has a capacity usable as a positive active material. Capacity also increases, so that the amount of a lithium ion increases especially, when the amount of a lithium metal is 1.05 mols or less. Therefore, it turns out that the amount of the optimal lithium ion is  $1.00 < x < 1.05$ .

[0032] Furthermore, the fixed quantity of the content of Li, nickel, and Co in the  $\text{Li}_x\text{Ni}_{0.7}\text{Co}_{0.302}$  active material manufactured according to the example 5 or the example 8 was carried out using ICP (inductively coupled plasma), and the result was shown in the following table 1.

[Table 1]

	Li[モル%]	Ni[モル%]	Co[モル%]	備考
実施例 5 ( $x=0.95$ )	6.52/6.94 = 0.94	40.9/58.71 = 0.70	17.77/58.93 = 0.30	$\text{Li}_{0.94}\text{Ni}_{0.70}\text{Co}_{0.30}\text{O}$
実施例 6 ( $x=1.00$ )	6.88/6.94 = 0.96	40.83/58.71 = 0.70	17.53/58.98 = 0.80	$\text{Li}_{0.96}\text{Ni}_{0.70}\text{Co}_{0.30}\text{O}$
実施例 7 ( $x=1.04$ )	6.92/6.94 = 0.997	40.36/58.71 = 0.69	17.47/58.93 = 0.30	$\text{Li}_{0.997}\text{Ni}_{0.69}\text{Co}_{0.30}\text{O}_2$
実施例 8 ( $x=1.06$ )	6.99/6.94 = 1.01	39.1/58.71 = 0.67	16.96/58.93 = 0.29	$\text{Li}_{1.01}\text{Ni}_{0.67}\text{Co}_{0.29}\text{O}_2$

It turns out that the amount of Li contained by the example 5 of this invention or 8 in the active material finally obtained when  $0.30\text{Li}_x\text{Ni}_{0.7}\text{Co}_2$  active material is manufactured changing the amount of a lithium to 0.95 or 1.06 mols is about 0.94 or about 1.06 mols as shown in Table 1.

[0033]

[Effect of the Invention] As mentioned above, this invention can manufacture the active material which excelled [ adjust / the lithium equivalent ] in an electrochemical performance and capacity. Moreover, in this invention, since the distribution of each constituent of the precursive inside of the body which is a reactant by using a macromolecule

as a chelating agent becomes uniform, an impurity phase is not generated at all in heat treatment process, but thereby, synthetic temperature can be made low or synthetic time can be shortened if needed.

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#### DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the graph which showed the X diffraction pattern of the positive active material for lithium secondary batteries manufactured according to the example of this invention.

[Drawing 2] It is the SEM photograph of the positive-active-material precursor for lithium secondary batteries manufactured while adjusting the equivalent of lithium salt according to the example of this invention.

[Drawing 3] It is the SEM photograph of the positive active material for lithium secondary batteries heat-treated and manufactured once, adjusting the equivalent of lithium salt according to the example of this invention.

[Drawing 4] It is the graph which showed the charge/discharge capability ability of the coin cell manufactured using the positive active material for lithium secondary batteries manufactured by heat-treating once according to the example of this invention.

[Drawing 5] It is the graph which showed the charge/discharge capability ability of the coin cell manufactured using the positive active material for lithium secondary batteries manufactured by heat-treating twice according to the example of this invention.

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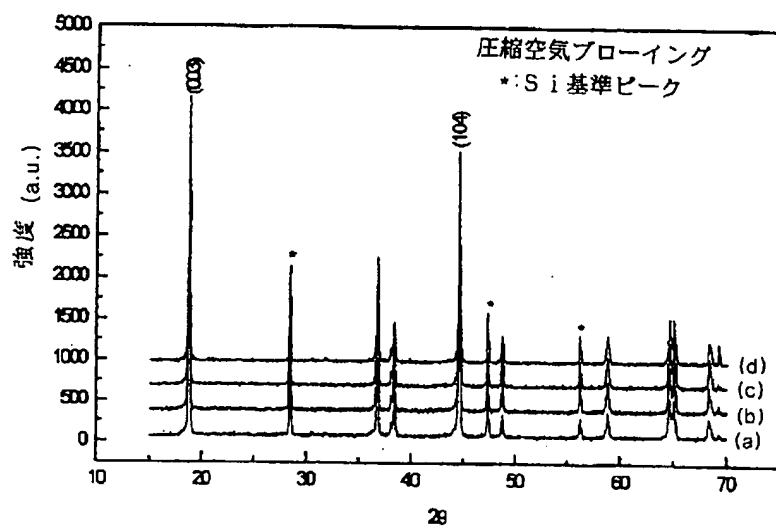
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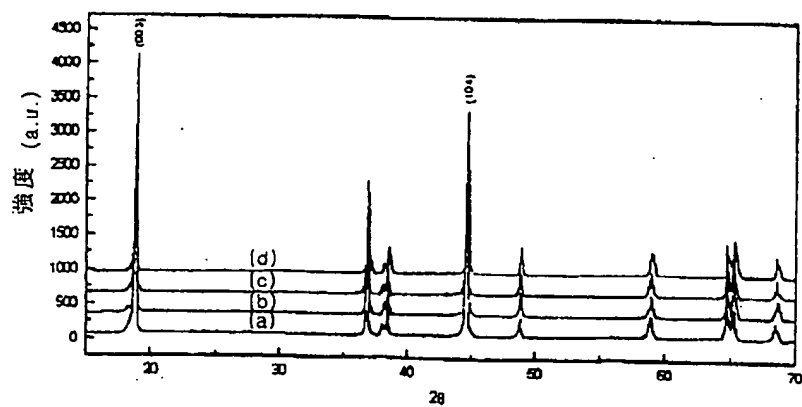
# DRAWINGS

[Drawing 1]

(a)



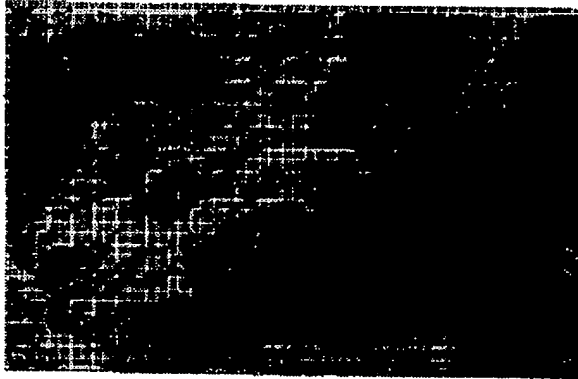
(b)



[Drawing 2]

図面代用写真

(a)



(b)



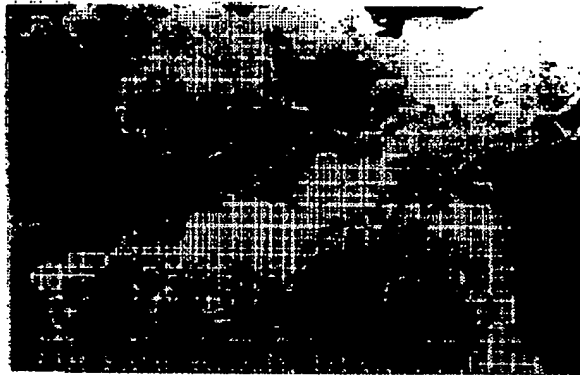
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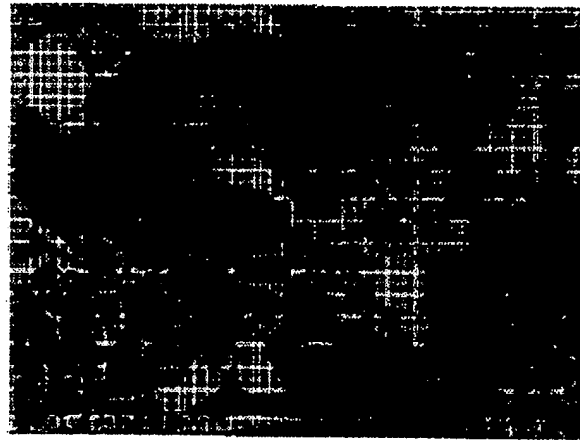
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図面代用写真

(a)



(b)

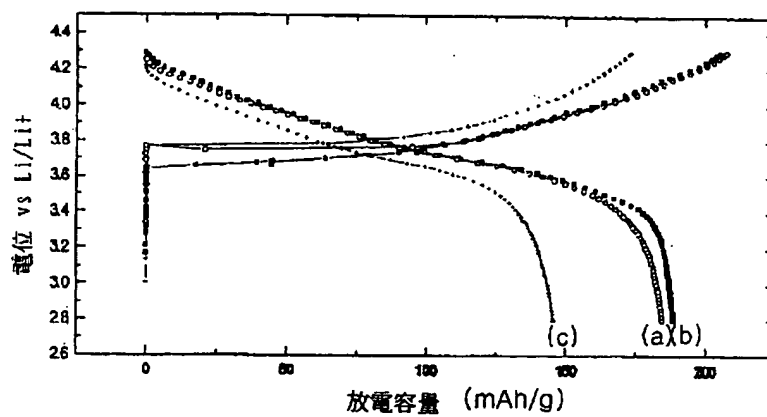


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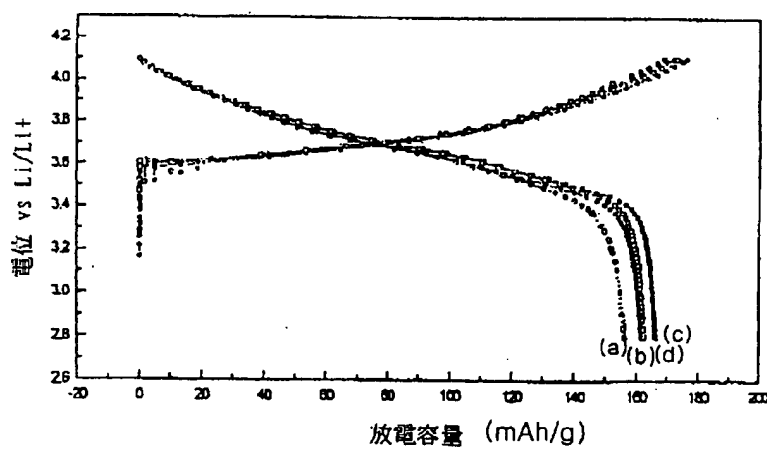


[Drawing 4]

(a)



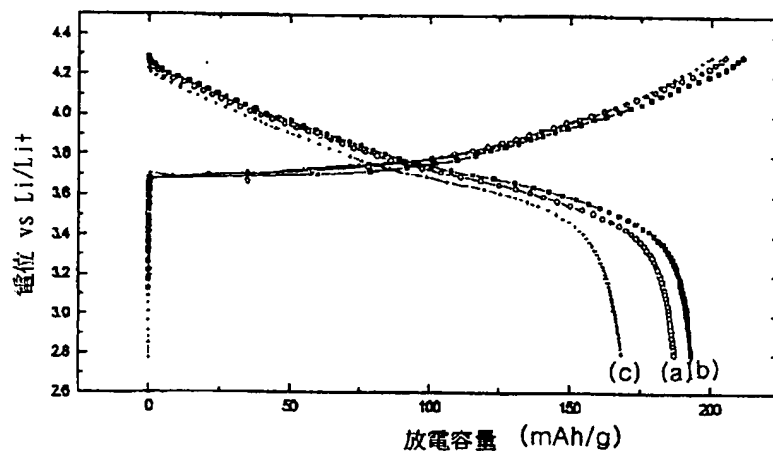
(b)



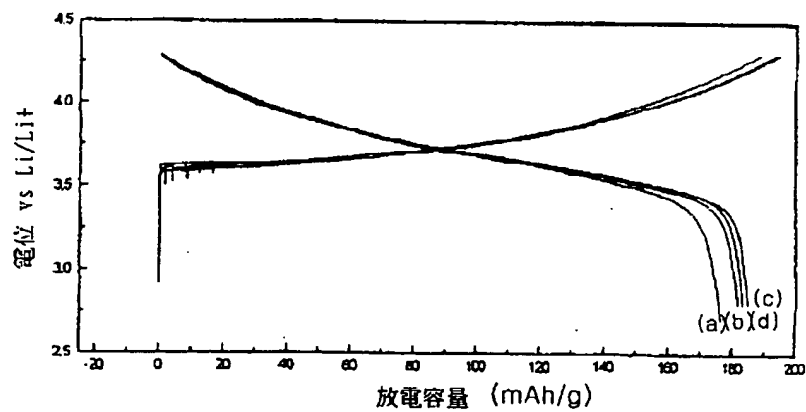
[Drawing 5]



(a)



(b)



[Translation done.]